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Classical and quantum pumping in closed systems

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Pumping of charge (Q) in a closed ring geometry is not quantized even in the strict adiabatic limit. The deviation form exact quantization can be related to the Thouless conductance. We use the Kubo formalism as a starting point for the calculation of both the dissipative and the adiabatic contributions to Q. As an application we bring examples for classical dissipative pumping, classical adiabatic pumping, and in particular we make an explicit calculation for quantum pumping in case of the simplest pumping device, which is a 3 site lattice model. We make a connection with the popular S matrix formalism which has been used to calculate pumping in open systems.

Pumping of charge in mesoscopic [3] and molecular size devices is regarded as a major issue in the realization of future "quantum circuits" or "quantum gates", possibly for the purpose of "quantum computing". Of particular interest is the possibility to realize a pumping cycle that transfers *exactly* one unit of charge per cycle [4, 5, 6, 7]. In open systems this "quantization" holds only approximately. But it has been argued [6] that the deviation from quantization is due to "dissipative" effect, and that exact quantization would hold in the strict adiabatic limit, if the system were *closed*. In this Letter we would like to show that the correct picture is quite different. In particular we would like to make a proper distinction between "dissipative" and "adiabatic" contributions to the pumping, and to calculate the deviation from exact quantization in the latter case. As a starting point we adopt the traditional Kubo formula [8], but we also point out the relation to the "adiabatic" [9, 10] and to the "S-matrix" [11] formulations. The present formulation of the pumping problem has few advantages: It is not restricted to the adiabatic regime; It give a "level by level" understanding of the pumping process; It allows the consideration of any type of occupation (not necessarily Fermi occupation); It allows future incorporation of external environmental influences such as that of noise; It regards the "voltage" over the pump as "electro motive force", rather than adopting the conceptually complicated view [12] of having a "chemical potential difference". Finally, on the practical level, we give a solution for the pumping in a 3 site lattice model. This is definitely the simplest pump circuit possible, and we believe that it can be realized as a molecular size device. It also can be regarded as an approximation for the closed geometry version of the two delta potential pump [7].

The structure of this Letter is as follows: We show how to get from the Kubo formalism an expression for the pumped charge Q, and explain the distinction between "dissipative" and the "adiabatic" contributions. Then we give illuminating examples for classical dissipative pumping and for classical adiabatic pumping. Next we discuss the case of quantum pumping, where the cycle is around a chain of degeneracies. We show that this can be understood as a special case of "adiabatic transfer" scheme. In order to get a quantitative estimate for the pumped charge we consider a 3 site lattice model. We get expressions for Q, and express them in terms of the Thouless conductance. We conclude by a short discussion of the relation between the Kubo formalism, the adiabatic formalism, and the S-matrix formalism.

Consider a system that has a ring geometry (Fig.1a). The Hamiltonian is $\mathcal{H}(x_1(t), x_2(t), x_3(t))$, where x_1 and x_2 are parameters that control the shape of the ring, or the height of some barriers, while $x_3{=}\Phi{=}\hbar\phi$ is the magnetic flux. We use units such that the elementary charge is unity. The "generalized forces" are conventionally defined as $F^k \equiv -\partial\mathcal{H}/\partial x_k$. In particular $\langle F^3\rangle$ is the current I through the ring (see remark [18]). Consider for a moment the time independent Hamiltonian $\mathcal{H}(x)$, with $x = \text{const}$, and assume that the system is prepared in a *stationary* state (either pure or mixed). The expectation value $\langle F^k\rangle$ of a generalized force is known as the "conservative force" or (in case of $k{=}3$) as the "persistent current". The "fluctuations" of the generalized forces are conventionally characterized by the real functions:

$$C^{ij}(\tau) \;=\; \langle \tfrac{1}{2}(F^i(\tau)F^j(0) + F^j(0)F^i(\tau))\rangle \qquad (1)$$

$$K^{ij}(\tau) \;=\; \frac{i}{\hbar}\langle [F^i(\tau), F^j(0)]\rangle \qquad (2)$$

Note that both functions have a well defined classical limit. Their Fourier transform will be denoted by $\tilde{C}^{ij}(\omega)$ and $\tilde{K}^{ij}(\omega)$ respectively.

Our interest in the following is in a *driving cycle*, where $x = x(t)$ forms a loop in the 3 dimensional parameter space. In linear response theory [8] the non-conservative contribution to $\langle F^k\rangle$ is related to $x(t)$ by a causal response kernel $\alpha^{ij}(t-t')$. The Kubo expression for this response kernel is $\alpha^{ij}(\tau) = \Theta(\tau)\; K^{ij}(\tau)$. Its Fourier transform is the generalized susceptibility $\chi^{ij}(\omega)$. From here we can derive the expression $\langle F^k\rangle = -\sum_j \mathbf{G}^{kj}\,\dot{x}_j$, where $\mathbf{G}^{kj}$ is the generalized conductance matrix:

$$\mathbf{G}^{ij} \;=\; \lim_{\omega\to 0}\frac{\mathrm{Im}[\chi^{ij}(\omega)]}{\omega} \;=\; \int_0^\infty K^{ij}(\tau)\tau d\tau \qquad (3)$$

Following Berry and Robbins [10] we split the conductance matrix into symmetric and anti-symmetric parts. Namely $\mathbf{G}^{ij} = \boldsymbol{\eta}^{ij} + \mathbf{B}^{ij}$. The antisymmetric part $\mathbf{B}$ can be regarded as a vector $\vec{\mathbf{B}} = (\mathbf{B}^{23}, \mathbf{B}^{31}, \mathbf{B}^{12})$, and the expression for the current can be written in an abstract way as $\langle F\rangle = -\boldsymbol{\eta}\cdot\dot{x} - \mathbf{B}\wedge\dot{x}$.

The rate of dissipation, which is defined as the rate in which energy is absorbed into the system, is given by $\dot{\mathcal{W}} = -\langle F\rangle\cdot\dot{x} = \sum_{kj} \boldsymbol{\eta}^{ij}\dot{x}_i\dot{x}_j$. Only the symmetric part of $\mathbf{G}^{ij}$ is responsible for dissipation of energy. The adiabatic regime is defined by the condition $|\dot{x}| \ll \Delta^2/\hbar\sigma$, where Δ is the typical level spacing, and σ is the root mean square value of the matrix element $(\partial\mathcal{H}/\partial x)_{nm}$ between neighboring levels. In the adiabatic regime $\boldsymbol{\eta}^{ij}$ vanishes because of the discreteness of the energy spectrum [10]. But outside of the adiabatic regime the levels acquires an effective width $\Gamma/\Delta = ((\hbar\sigma/\Delta^2)V)^{2/3} > 1$ and therefore the smoothed version of $\tilde{K}^{ij}(\omega)$ should be considered. Consequently one can obtain the fluctuation-dissipation (FD) relation: $\boldsymbol{\eta}^{ij} \sim \tilde{C}^{ij}(\omega=0)$. The formulation of the exact FD relation depends on the assumptions regarding the occupation $f(E_n)$ of the energy levels. See [13, 14]. Commonly one assumes a zero temperature Fermi occupation, but this is not essential for the following analysis. In order to derive the above expression for Γ we have used the result of [13] (Sec.17) for the "core width" at the breaktime $t = t_{\rm prt}$ of perturbation theory. Note that in the semiclassical limit (small $\hbar$) the adiabaticity condition always breaks down.

The antisymmetric part $\mathbf{B}$ of $\mathbf{G}^{ij}$ does not have to vanish in the adiabatic limit. It can be obtained from the adiabatic equation by looking for a first-order stationary-like solution [4, 5, 10], but we prefer to regard it as a term in the (full) Kubo expression Eq.(3). In [9, 10] it has been demonstrated that it can be written as

$$\begin{aligned}\mathbf{B}^{ij} &= -2\hbar\sum_n f(E_n)\ \mathrm{Im}\left\langle \frac{\partial}{\partial x_i}n(x)\middle|\frac{\partial}{\partial x_j}n(x)\right\rangle \\ &= 2\hbar\sum_{m\neq n} f(E_n)\frac{\mathrm{Im}\left[\left(\frac{\partial\mathcal{H}}{\partial x_i}\right)_{nm}\left(\frac{\partial\mathcal{H}}{\partial x_j}\right)_{mn}\right]}{(E_m-E_n)^2} \qquad (4)\end{aligned}$$

Note that the "vertical" component of $\vec{\mathbf{B}}$ vanishes in the "horizontal" x_3=0 plane due to time reversal symmetry.

Disregarding a possible persistent current contribution (that does not exist in the case of a planar Φ=0 cycle), the expression for the pumped charge is:

$$Q = \oint I dt = -\left[\oint \boldsymbol{\eta}\cdot dx + \oint \mathbf{B}\wedge dx\right]_{k=3} \qquad (5)$$

If we neglect the first term, which is associated with the dissipation effect, and average the second ("adiabatic") term over the flux, then we get

$$\overline{Q|_{\rm adiabatic}} = -\frac{1}{2\pi\hbar}\iint \mathbf{B}\cdot\vec{dx}\wedge\vec{dx} = \text{integer} \qquad (6)$$

The integration should be taken over a cylinder of vertical height $2\pi\hbar$, and whose basis is determined by the projection of the pumping cycle onto the (x_1, x_2) plane. The last equality is argued as follows: The flux $(1/\hbar)\iint \mathbf{B}\cdot dx\wedge dx$ through a surface that is enclosed by a cycle is the Berry phase [9]. The result should be independent of the surface. Therefore the flux through a *closed* surface should equal $2\pi\times$integer. Integrating over a cylinder, as in Eq.(6), is effectively like integrating over a closed surface (because of the 2π periodicity in the vertical direction). This means that the flux averaged Q of Eq.(6) has to be an integer.

Before we discuss the quantum mechanical pumping, it is instructive to bring two simple examples for *classical* pumping. In the following we consider one particle ($\mathbf{r}$) in a two dimensional ring as in Fig.1a.

The first example is for classical *dissipative* pumping. The conductance $G = \mathbf{G}^{33}$ can be calculated for this system [14] leading to a mesoscopic variation of the Drude formula. The current is $I = -G\times\dot{\Phi}$, where $-\dot{\Phi}$ is the electro-motive-force. Consider now the following pumping cycle: Change the flux from Φ_1 to Φ_2, hence pumping charge $Q = -G(1)\times(\Phi_2-\Phi_1)$. Change the conductance from $G(1)$ to $G(2)$ by modifying the shape of the ring. Change the flux from Φ_2 back to Φ_1, hence pumping charge $Q(2) = -G(2)\times(\Phi_1-\Phi_2)$. Consequently the net pumping is $Q = (G(2)-G(1))\times(\Phi_2-\Phi_1)$.

The second example is for classical *adiabatic* pumping. The idea is to trap the particle inside the ring by a potential well $U_{\rm trap}(\mathbf{r}_1-x_1(t), \mathbf{r}_2-x_2(t))$. Then make a translation of the trap along a circle of radius R, namely $x(t) = (R\cos(\Omega t), R\sin(\Omega t), \Phi{=}\text{const})$. It is a-priori clear that in this example the pumped charge per cycle is $Q = 1$, irrespective of Φ. Therefore the $\vec{\mathbf{B}}$ field must be

$$\vec{\mathbf{B}} = -\frac{(x_1, x_2, 0)}{2\pi(x_1^2+x_2^2)} \qquad (7)$$

This can be verified by calculation via Eq.(4). The singularity along the x_3 axis is not of quantum mechanical origin: It is not due to degeneracies, but rather due to the diverging current operator ($\partial\mathcal{H}/\partial x_3 \propto 1/\sqrt{x_1^2+x_2^2}$).

We turn now to the quantum mechanical case. Consider an adiabatic cycle that involves a particular energy level n. This level is assumed to have a degeneracy point at $(x_1^{(0)}, x_2^{(0)}, \Phi^{(0)})$. It follows that in fact there is a vertical "chain" of degeneracy points that are located at $(x_1^{(0)}, x_2^{(0)}, \Phi^{(0)} + 2\pi\hbar\times\text{integer})$. These degeneracy points are important for the geometrical understanding of the $\mathbf{B}$ field, as implied by Eq.(4). Every degeneracy point is like a monopole charge. The total flux that emerges from each monopole must be $2\pi\hbar\times$integer for a reason that was explained after Eq.(6). Thus the monopoles are quantized in units of $\hbar/2$.

The $\mathbf{B}$ field which is created (so to say) by a vertical chain of monopoles may have a different "near field" and "far field" behavior, which we discuss below. (Later we further explain that "near field" means regions in x space, in the vicinity of degeneracy points, where $g_T \gg 1$, while "far field" means regions where $g_T \ll 1$). The far field regions exist if the chains are well isolated. The far field region of a given chain is obtained by regarding the chain as a smooth line. This leads *qualitatively* to the same field as in Eq.(7). Consequently, for a "large radius"

pumping cycle in the $\Phi = 0$ plane, we get $|Q| \approx 1$. In the following we are interested in the deviation from "exact" quantization: If $\phi^{(0)} = 0$ we expect to have $|Q| \geq 1$, while if $\phi^{(0)} = \pi$ we expect $|Q| \leq 1$. Only for the ϕ averaged Q of Eq.(6) we get *exact quantization*.

The deviation from $|Q| \approx 1$ is extremely large if we consider a tight pumping cycle around a $\phi^{(0)} = 0$ degeneracy. After linear transformation of the shape parameters, the energy splitting $\Delta = E_n - E_m$ of the energy level n from its neighboring (nearly degenerated) level m can be written as $\Delta = ((x_1 - x_1^{(0)})^2 + (x_2 - x_2^{(0)})^2 + c^2(\phi - \phi^{(0)})^2)^{1/2}$ where c is a constant. The monopole field is accordingly

$$\vec{\mathbf{B}} = \pm\frac{c}{2}\,\frac{(x_1 - x_1^{(0)}, x_2 - x_2^{(0)}, x_3 - x_3^{(0)})}{((x_1 - x_1^{(0)})^2 + (x_2 - x_2^{(0)})^2 + (\frac{c}{\hbar})^2(x_3 - x_3^{(0)})^2)^{3/2}} \quad (8)$$

where the prefactor is determined by the requirement of having a single $(\hbar/2)$ monopole charge. Assuming a pumping cycle of radius R in the $\Phi = 0$ plane we get from the second term of Eq.(5) that the pumped charge is $Q = \mp\pi\sqrt{g_T}$, where $g_T = (\partial^2\Delta/\partial\phi^2)/\Delta = c^2/R^2$ is a practical definition for the Thouless conductance in this context. It is used here simply as a measure for the sensitivity of an energy level to the magnetic flux Φ.

What we want to do in the following is to "interpolate" between the "near field" result, which is $Q = \mathcal{O}(\sqrt{g_T})$, and the "far field" result, which is $Q = \mathcal{O}(1)$. For this purpose it is convenient to consider a particular model that can be solved exactly. We consider a ring with two barriers. The model is illustrated in Fig.2. A version of this model, where the two barriers are modeled as "delta functions", has been analyzed in [7] in case of *open* geometry. Below we are going to analyze a different version of the two barrier model, that allows an exact solution for *closed* geometry.

We can classify the eigenstates of the *closed* ring into two categories: wire states, and dot states (Fig.2a). The latter are those states that are localized in the "dot region" in the limit of infinitely high barriers. In case of zero temperature Fermi occupation we define E_F as the energy of the last occupied wire level in the limit of infinitely high barriers. The two "shape" parameters are the the bias x_1, and the dot potential x_2. The bias determines whether the dot tends to exchange particles via the *left* or via the *right* barrier. The dot potential is loosely defined as the energy of the dot level (Fig.2a). A model specific definition of these parameters in the context of the 3-site lattice Hamiltonian will be given later.

The pumping cycle is assumed to be in the $\Phi = 0$ plane, so there is no issue of "conservative" persistent current contribution. We start with a positive bias ($x_1 > 0$) and lower the dot potential from a large $x_2 > E_F$ value to a small $x_2 < E_F$ value. As a result, one electron is transfered via the *left* barrier into the dot region. Then we invert the bias ($x_1 < 0$) and raise back x_2. As a result the electron is transfered back into the wire via the *right* barrier. A closer look at the above scenario (Fig.2b) reveals the following: As we lower the dot potential across a wire level, an electron is adiabatically transfered once from left to right and then from right to left. As long as the bias is positive ($x_1 > 0$) the net charge being pumped is very small ($|Q| \ll 1$). Only the lowest wire level that participate in the pumping cycle carries $Q = \mathcal{O}(1)$ net charge: It takes an electron from the left side, and after the bias reversal it emits it into the right side. Thus the pumping process in this model can be regarded as a particular example [5] of an *adiabatic transfer scheme*: The electrons are adiabatically transfered from state to state, one by one, as in "musical chair game".

For a single occupied level the net Q is the sum of charge transfer events that take place in few avoided crossings. For many particle occupation the total Q is the sum over the net Qs which are carried by individual levels. For a dense zero temperature Fermi occupation the summation over all the net Qs is a telescopic sum, leaving non-canceling contributions only from the first and the last adiabatic crossings. The latter involve the last occupied level at the Fermi energy.

In order to get a quantitative estimate for the Q in a given avoided crossing, we consider the simplest version of the "two barrier model" that still contains all the essential ingredients: This is a three site lattice system. The middle site supports a single "dot state", while the two other sites support two "wire states". The Hamiltonian is

$$\mathcal{H} \mapsto \begin{pmatrix} 0 & c_1 & \mathrm{e}^{i\phi} \\ c_1 & u & c_2 \\ \mathrm{e}^{-i\phi} & c_2 & 0 \end{pmatrix} \quad (9)$$

The three parameters are the bias $x_1 = c_1 - c_2$, the dot potential $x_2 = u$, and the flux $x_3 = \Phi = \hbar\phi$. For presentation purpose we assume that $0 < c_1, c_2 \ll 1$. The eigenstates are E_n. Disregarding the coupling between the "wires" and the "dot" we have two wire states with $E = \pm 1$, and a dot state with $E = u$. Taking into account the wire-dot coupling we find that there are two vertical chains of degeneracies. The $u \approx -1$ chain is $(0, -1 + c_1^2, 2\pi\hbar \times \text{integer})$ and the $u \approx 1$ chain is $(0, +1 + c_1^2, \pi + 2\pi\hbar \times \text{integer})$.

The eigenvalues E_n are the solutions of a cubic equation. Rather than writing the (lengthy) analytical expressions for them we give a numerical example for their dependence on u in the inset of Fig.3. The eigenstates are

$$|n(x)\rangle \mapsto \frac{1}{\sqrt{S}} \begin{pmatrix} c_2\mathrm{e}^{i\phi} + c_1 E_n \\ 1 - E_n^2 \\ c_1\mathrm{e}^{-i\phi} + c_2 E_n \end{pmatrix} \quad (10)$$

where S is the normalization. Note that for $E = \pm 1$ we have $S = 2(c_1 \pm c_2)^2$, while for $E = 0$ we have $S \approx 1$. After some algebra we find that the first component of the $\vec{\mathbf{B}}$ field in the $\Phi = 0$ plane is

$$\mathbf{B}^1 = -2\mathrm{Im}\left\langle \frac{\partial}{\partial u} n(x) \middle| \frac{\partial}{\partial \phi} n(x) \right\rangle = -(c_1^2 - c_2^2)\frac{1}{S^2}\frac{\partial S}{\partial u}$$

Which is illustrated in Fig.3. For a pumping cycle around the $u \approx \mp 1$ vertical "chain" the main contribution to Q comes from crossing the $u \approx \mp 1$ line. Hence we get

$$Q = \pm \frac{c_1 \pm c_2}{c_1 \mp c_2} = \pm \sqrt{1 \pm 2g_T} \tag{11}$$

where the Thouless conductance in this context is defined as $g_T = 2c_1c_2/(c_1 \mp c_2)^2$. In both cases we have approximate quantization $Q = \pm 1 + \mathcal{O}(g_T)$ for $g_T \ll 1$, while for a tight cycle either $Q \to \infty$ or $Q \to 0$ depending on which line of degeneracies is being encircled. If the pumping cycle encircles both "chains" then we get $Q = 4c_1c_2/(c_1^2 - c_2^2)$. In the latter case $Q = \mathcal{O}(g_T)$ for $g_T \ll 1$, with no indication for quantization.

For a pumping in a dot-wire system (see illustration in Fig.1b), in the limit of a very long wire (many sites) we express the Kubo formula for the conductance matrix using the S matrix of the dot region. The derivation assumes "quantum chaos", and leads to

$$\mathbf{G}^{3j} = \frac{1}{2\pi i}\mathrm{trace}\left(P\frac{\partial S}{\partial x_j}S^\dagger\right) \tag{12}$$

This is easily identified as the Büttiker-Prétre-Thomas formula [11], which has been derived for quantum pumping in open systems (Fig.1e). In particular we get $G^{33} = (1/(2\pi\hbar))\mathrm{trace}(PS(1-P)S^\dagger)$, which is just the Landauer formula [15, 16, 17].

In summary we have shown how the Kubo formalism can be used in order to derive both classical and quantum mechanical results for the pumped charge Q in a closed system. In this formulation the distinction between dissipative and non-dissipative contributions is manifest. The dissipative contribution to the pumping can be neglected in the adiabatic regime. However, if the adiabaticity condition is violated it does not mean automatically that we have a dissipative effect. Classical pumping by translation is an obvious example. For the derivation of the dissipative part of the Kubo formula it is essential to realize that in generic circumstances (unlike the case of translations) the adiabatic equation does not possess a stationary solution.

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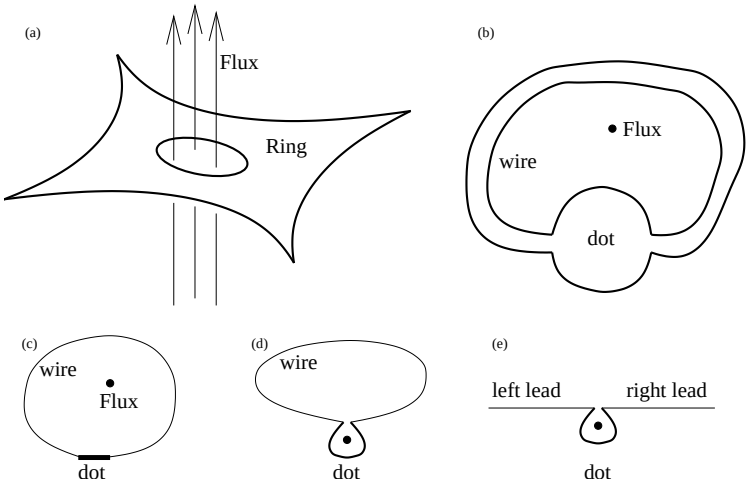

FIG1. Illustration of a ring system (a). The shape of the ring is controlled by some parameters x_1 and x_2. The flux through the ring is $x_3 = \Phi$. A system with equivalent topology, and abstraction of the model are presented in (b) and (c). The "dot" can be represented by an S matrix that depends on x_1 and x_2. In (d) also the flux x_3 is regarded as a parameter of the dot. If we "cut" the wire in (d) we get the open two lead geometry of (e).

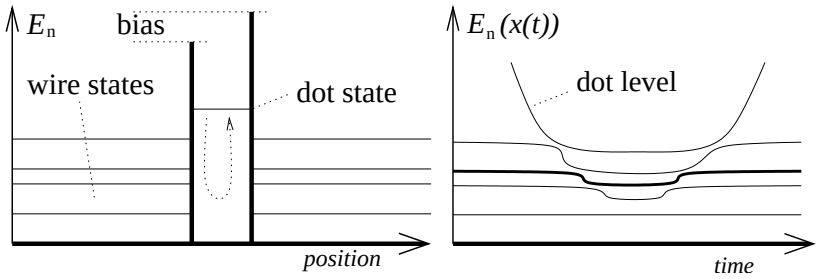

FIG2. Schematic illustration of quantum pumping in a closed wire-dot system. The net charge via the third level (thick solid line on the right) is vanishingly small: As the dot potential is lowered an electron is taken from the left side (first avoided crossing), and then emitted back to the left side (second avoided crossing). Assuming that the bias is inverted before the dot potential is raised back, only the second level carry a net charge $Q = \mathcal{O}(1)$.

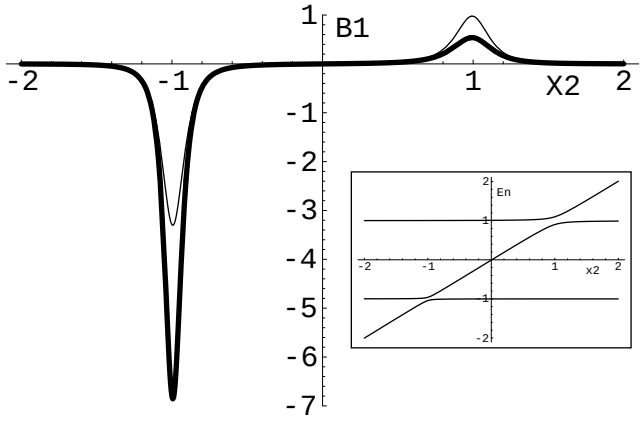

FIG3. The first component of the **B** field for a particle in the middle level of the 3 site lattice model. It is plotted as a function of the dot potential $x_2 = u$. The other parameters are $\phi = 0$, and $c_1 = 0.1$, while $c_2 = 0.04$ for the thick line and $c_2 = 0.02$ for the thin line. In the limit $c_2 \to 0$, all the charge that is transfered from the left side into the dot during the first avoided crossing, is emitted back into the left side during the second avoided crossing. Inset: The eigenenergies $E_n(x)$ for the $c_2 = 0.04$ calculation.